Electronic Appendix A: A sample calculation of the trace-element evolution of metasomatic liquid and cumulates during the differentiation of metasomatic melt within the lithosphere

Here we provide an example of a Monte Carlo simulation following the model presented in Fig. 1 (here, and throughout this Appendix, references to specific figures and tables are to those in the main article). As shown in Fig. 1, the model is divided into two parts:
(1) Generation of a metasomatic liquid (liq) by low degrees of melting of a source that varies in composition from E-DMM to DMM;
(2) Differentiation of this low-degree melt and the formation of anhydrous and hydrous cumulates.

In practice, the calculation follows the schematic model of vein formation illustrated in Fig. 4 of the article. First, we calculate the composition of the initial metasomatic liquid (L_0), then the anhydrous cumulate composition as well as the composition of liquid L_1, and finally the composition of hydrous cumulates and the residual liquid L_2.

In the following example of the calculation, the parameters that are allowed to vary are shown in yellow boxes, parameters that are held constant are not highlighted, while the calculated values are listed in gray boxes.

Part 1 - Composition of the initial metasomatic liquid L_0

The composition of the initial metasomatic liquid L_0 depends on the source composition, the degree of partial melting, the solidus phase proportions, and the proportions of phases that enter the melt.

The initial mantle source composition was calculated by varying the proportion of E-DMM to DMM in the source along with the trace-element compositions of these two end members (Workman and Hart, 2005):

0.519 Fraction of E-DMM in the source (can vary from 0 to 1); in this example, the fraction of DMM is 0.481

Composition of E-DMM and DMM end members (Workman and Hart, 2005):

	Rb	Ba	Th	U	Nb	La	Ce	Pb	Pr	Sr	Nd	Sm	Zr	Hf	Eu	Ti	Gd	Tb	Dy	Ho	Y	Er	Yb	Lu
E-DMM (ppm)	0.11	1.22	0.016	0.005	0.25	0.25	0.73	0.024	0.13	9.72	0.70	0.27	6.09	0.19	0.11	792	0.40	0.076	0.54	0.123	3.55	0.37	0.38	0.060
DMM (ppm)	0.05	0.56	0.008	0.003	0.15	0.19	0.55	0.018	0.11	7.66	0.58	0.24	5.08	0.16	0.10	716	0.36	0.070	0.51	0.115	3.33	0.35	0.37	0.058

Calculated source:

	Rb	Ba	Th	U	Nb	La	Ce	Pb	Pr	Sr	Nd	Sm	Zr	Hf	Eu	Ti	Gd	Tb	Dy	Ho	Y	Er	Yb	Lu
Source (ppm)	0.08	0.90	0.012	0.004	0.20	0.22	0.64	0.021	0.12	8.73	0.64	0.26	5.60	0.17	0.10	756	0.38	0.073	0.52	0.119	3.44	0.36	0.374	0.059

The initial metasomatic liquid (shown in Fig. 5a as L_0) is calculated using the non-modal aggregate fractional melting equation $[C_l/C_o=(1/F)(1-(1-PF/D)^{(1/P)})]$ where C_l = the aggregated liquid, C_o = the source composition, F = the melt fraction, P = the bulk distribution coefficient for those phases entering the melt, and D = the bulk distribution coefficient at the beginning of melting; the source mineral modes are from Table 1 and melting modes are from Walter (1998)

and are for partial melting at 3 GPa (they have been slightly modified to take into account the addition of minor sulphide in the source; Table 1). The $D^{min/liq}$s for olivine (ol), orthopyroxene (opx), clinopyroxene (cpx), garnet (gt), and sulphide are reported in Table 2. The degree of partial melting is a free parameter that can vary between 0.5% and 1.8 % according the distribution shown in Fig. 2b.

0.0077 Degree of partial melting (can vary from 0.005 to 0.018).

Calculated bulk distribution coefficients D and P:

	Rb	Ba	Th	U	Nb	La	Ce	Pb	Pr	Sr	Nd	Sm	Zr	Hf	Eu	Ti	Gd	Tb	Dy	Ho	Y	Er	Yb	Lu
D	0.0004	0.0005	0.0003	0.0009	0.0069	0.012	0.019	0.006	0.025	0.026	0.047	0.081	0.065	0.088	0.135	0.110	0.148	0.175	0.202	0.248	0.308	0.398	0.517	0.675
P	0.0007	0.0006	0.0011	0.0021	0.0120	0.046	0.075	0.052	0.093	0.104	0.176	0.297	0.193	0.300	0.493	0.366	0.532	0.606	0.666	0.787	0.943	1.195	1.532	1.967

Composition of the initial metasomatic liquid (corresponding to liquid L_0 plotted in Fig. 5a) using F= 0.0077 and the mantle source composition calculated above:

	Rb	Ba	Th	U	Nb	La	Ce	Pb	Pr	Sr	Nd	Sm	Zr	Hf	Eu	Ti	Gd	Tb	Dy	Ho	Y	Er	Yb	Lu
L0 (ppm)	10.46	117.9	1.56	0.55	17.48	14.02	27.59	2.05	4.17	291.6	12.87	3.06	82.4	1.91	0.75	6719	2.52	0.41	2.59	0.48	11.19	0.90	0.73	0.09

Part 2 – Composition of model anhydrous and model hydrous cumulates

1) Composition of anhydrous cumulates and residual liquid L_1

The compositions of the anhydrous cumulates and the residual liquid (reported as L_1 in Fig. 5b) are calculated using the fractional crystallization equation, the composition of the initial metasomatic liquid (L_0), $D^{min/liq}$s reported in Table 2, and proportions of clinopyroxene, garnet, olivine, orthopyroxene, and trapped liquid that are allowed to vary within bounds that are constrained by experimental petrology and the modes of xenolithic metasomatic vein material.

Mineralogical composition of the anhydrous fractionating assemblage:

garnet	0.122	Free to vary between 0 and 0.3
olivine	0.043	With garnet = 0.122, olivine can vary between 0.028 and 0.066†
opx	0.008	Free to vary between 0 and 0.08
trapped liq.	0.045	Free to vary between 0 and 0.05
cpx	0.781	Fraction of cpx in the cumulate assemblage fixed by the expression: 1 – sum of all other phases

† High-pressure experiments performed between 1.5 and 1 GPa suggest that the anhydrous cumulate assemblage formed at such pressures is clinopyroxene + minor olivine (Nekvasil *et al.*, 2004; Pilet *et al.*, 2010). However, experiments at higher pressures indicate that the cumulate assemblage that crystallizes between ~3 and 2.5 GPa is clinopyroxene + garnet (Hauri *et al.*, 1994). Therefore, as indicated in Fig. 4, we include a link between garnet and olivine abundances in the anhydrous cumulate assemblage to mimic this pressure effect.

Proportion of residual liquid ($f L_1$) after anhydrous cumulate fractionation: 0.595 This parameter is free to vary between 0.65 to 0.45.

Bulk distribution coefficients (D) for the anhydrous cumulate assemblage. The individual mineral-melt Ds are reported in Table 2:

	Rb	Ba	Th	U	Nb	La	Ce	Pb	Pr	Sr	Nd	Sm	Zr	Hf	Eu	Ti	Gd	Tb	Dy	Ho	Y	Er	Yb	Lu
Bulk D	0.046	0.046	0.046	0.046	0.054	0.088	0.115	0.053	0.137	0.146	0.202	0.299	0.181	0.285	0.459	0.371	0.481	0.512	0.520	0.561	0.616	0.715	0.875	1.057

Composition of the residual liquid after anhydrous cumulate fractionation (Liquid L_1 in Fig. 5b). The trace-element composition of this liquid is calculated using the fractional crystallization equation, $C_l/C_o=F^{(D-1)}$:

	Rb	Ba	Th	U	Nb	La	Ce	Pb	Pr	Sr	Nd	Sm	Zr	Hf	Eu	Ti	Gd	Tb	Dy	Ho	Y	Er	Yb	Lu
L1 (ppm)	17.16	193.57	2.56	0.91	28.57	22.51	43.69	3.35	6.53	454.3	19.48	4.40	126.2	2.76	0.99	9316	3.30	0.53	3.32	0.60	13.65	1.05	0.77	0.09

Composition of the model anhydrous cumulates (corresponding to the model anhydrous composition plotted in Fig. 5c):

	Rb	Ba	Th	U	Nb	La	Ce	Pb	Pr	Sr	Nd	Sm	Zr	Hf	Eu	Ti	Gd	Tb	Dy	Ho	Y	Er	Yb	Lu
Model anhydrous cumulate (ppm)	0.61	6.84	0.09	0.03	1.19	1.55	3.94	0.14	0.71	52.6	3.17	1.09	18.25	0.65	0.39	2905	1.37	0.24	1.51	0.30	7.56	0.69	0.65	0.09

2) Composition of the model hydrous cumulate assemblage and the residual liquid L_2

While the fractionation of hydrous cumulates is carried out between $f L_1$ (the end point of anhydrous fractionation) and $f L_2$, the plotted composition of the hydrous cumulates (e.g., Fig. 5c) is calculated between $f$max and $f L_2$; both $f$max and $f L_2$ are free variables (with constraints).

***f* max**	0.409	Free to vary between f after anhydrous cumulate formation (i.e. $f L_1$, in this example 0.595) and this value minus 0.3.
***f* L_2**	0.266	Free to vary between ($f$max – 0.1) and 0.2 with the condition that $f$max – $f L_2$ >0.1.

For the formation of the hydrous cumulates, the fractionating assemblage was separated into two parts (#1 and #2) in order that plagioclase and zircon crystallization be limited to the most differentiated and lowest temperature liquids. The transition between these two slightly different mineral assemblages was fixed at f = 0.4 (note that in some cases, $f L_2$ was higher that 0.4, so the second mineral assemblage was not used in the calculation).

Hydrous fractional assemblage (#1) between $f L_1$ and f = 0.4 (see Fig. 4):

cpx	0.198	Free to vary between 0 and 0.5
garnet	0.024	Free to vary between 0 and 0.03
rutile	0.0056	Free to vary between 0 and 0.01; rutile plus ilmenite is limited to a maximum of 0.01
ilmenite	0.0034	Ilmenite is free to vary from 0 to (0.01 – proportion rutile); in this example ilmenite can varied between 0 and 0.0044
sphene	0.0081	Free to vary between 0 and 0.02
allanite	0.00039	Free to vary between 0 and 0.0005
apatite	0.00281	Free to vary between 0 and 0.005
sulphide	0.00001	Free to vary between 0 and 0.001
opx	0.017	Free to vary between 0 and 0.02
phlogopite	0.060	Free to vary between 0 and 0.10*
amphibole	0.681	Fraction of amphibole in the cumulate assemblage fixed by the expression: 1 – sum of all other phases
total	1.000	

*As indicated in the main text, the model includes the addition of phlogopite associated with trapped liquid to simulate the modal and cryptic metasomatism observed at the periphery of metasomatic veins (see Fig. 4b). The proportion of phlogopite (representing veinlets in Fig. 4b) relative to trapped liquid (representing interstitial glass and metasomatism in the reaction zone, Fig. 4b) is assumed to be 1: 3 (i.e., the proportion of trapped liquid equal three times the proportion of phlogopite in the hydrous vein).

Hydrous fractional assemblage (#2) between f = 0.4 and f = L_2:

Cpx	0.4131	Free to vary between 0 and 0.5
rutile	0.0027	Free to vary between 0 and 0.01; rutile plus ilmenite is limited to a maximum of 0.01
ilmenite	0.00370	Ilmenite is free to vary from 0 to (0.01 – proportion rutile); in this example ilmenite can varied between 0 and 0.0073
sphene	0.0088	Free to vary between 0 and 0.02
apatite	0.0080	Free to vary between 0 and 0.018
zircon	0.0001	Free to vary between 0 and 0.0005
allanite	0.0002	Free to vary between 0 and 0.0005
sulfide	0.0034	Free to vary between 0 and 0.006
plagioclase	0.0062	Free to vary between 0 and 0.02
phlogopite	0.0601	Same constraints on phlogopite as in the hydrous fractionating assemble #1
amphibole	0.4939	Fraction of amphibole in the cumulate assemblage is fixed by the expression: 1 – sum of all other phases
total	1.0000	

a) Composition of the liquid at $f$max (from f = 0.595 to $f$max)

In the present example, $f$max is higher that 0.4 but lower than f after the fractionation of the anhydrous cumulates (fL_1). So, the first step is to calculate the composition of the residual liquid at $f$max using hydrous assemblage #1.

The bulk Ds are calculated with the phase proportions from hydrous assemblage #1 and the $D^{min/liq}$s reported in Table 2:

	Rb	Ba	Th	U	Nb	La	Ce	Pb	Pr	Sr	Nd	Sm	Zr	Hf	Eu	Ti *	Gd	Tb	Dy	Ho	Y	Er	Yb	Lu
Bulk D	0.247	0.484	0.441	0.124	1.07	0.93	0.98	0.09	1.19	0.53	1.44	1.77	0.40	0.66	1.69	2.00	1.82	1.75	1.68	1.62	1.56	1.43	1.22	1.19

* For Ti, the bulk D corresponds to an assemblage without rutile, ilmenite, and sphene. For these three phases, stoichiometric TiO_2 contents are used to calculate the effects of fractionation on liquid Ti contents (see discussion below).

Composition of the residual liquid at $f$max (f = 0.409):

	Rb	Ba	Th	U	Nb	La	Ce	Pb	Pr	Sr	Nd	Sm	Zr	Hf	Eu	Ti **	Gd	Tb	Dy	Ho	Y	Er	Yb	Lu
Residual liquid at $f$max (ppm)	22.8	235.0	3.16	1.26	27.8	23.1	44.0	4.7	6.1	542	16.5	3.3	158	3.1	0.76	5134	2.42	0.40	2.57	0.48	11.1	0.89	0.71	0.08

** The Ti content of the residual liquid was calculated in two steps. First, the Ti content of rutile, ilmenite and sphene weighted by their phase proportions was subtract from the initial liquid composition; second, the fractional crystallisation equation was used along with the remaining phases and their Ti D's to calculate the final TiO_2 content of the residual liquid.

b) Model hydrous cumulate and residual liquid compositions produced at f = 0.4

The bulk distribution coefficients are the same as those used for the calculation of the residual liquid at $f$max (the fractionating assemblage is the same).

Composition of residual liquid at f = 0.4:

	Rb	Ba	Th	U	Nb	La	Ce	Pb	Pr	Sr	Nd	Sm	Zr	Hf	Eu	Ti **	Gd	Tb	Dy	Ho	Y	Er	Yb	Lu
Residual liquid at f =0.4 (ppm)	23.1	237.6	3.20	1.28	27.8	23.2	44.0	4.8	6.06	547	16.3	3.2	160	3.2	0.75	4899	2.38	0.40	2.53	0.47	10.9	0.88	0.71	0.08

Hydrous cumulate composition produced between $f$max and 0.4 (in this example $f$max = 0.409):

	Rb	Ba	Th	U	Nb	La	Ce	Pb	Pr	Sr	Nd	Sm	Zr	Hf	Eu	Ti	Gd	Tb	Dy	Ho	Y	Er	Yb	Lu
Model hydrous cumulate (ppm)	5.67	114.3	1.40	0.16	29.8	21.46	43.1	0.41	7.21	289	23.7	5.79	63.8	2.07	1.28	15933	4.37	0.70	4.29	0.77	17.17	1.27	0.87	0.09

The composition of amphibole present in the cumulate assemblage is calculated using the following equation: $C^{amph} = C^{cumulate}/[\%amph + (\%min\_1 \times D^{min\_1/liq}/D^{amph/liq}) + \ldots + (\%min\_n \times D^{min\_n/liq}/D^{amph/liq})]$ where %min_1 to %min_n are the proportions of minerals 1 to n in the cumulate assemblage and $D^{min\_1/liq}$ to $D^{min\_n/liq}$ are the corresponding mineral-liquid distribution coefficients.

Amphibole composition in the model hydrous cumulate assemblage produced between $f$max and f = 0.4:

	Rb	Ba	Th	U	Nb	La	Ce	Pb	Pr	Sr	Nd	Sm	Zr	Hf	Eu	Ti[#]	Gd	Tb	Dy	Ho	Y	Er	Yb	Lu
Amphibole (ppm)	2.24	84.3	0.06	0.02	14.7	5.33	17.6	0.46	3.82	374	15.0	4.53	68.7	2.29	1.09	12272	3.96	0.65	4.03	0.73	16.3	1.19	0.78	0.09

[#] Ti in amphibole is calculated using the same equation as described previously, but taking into account the presence of cumulate Ti-rich phases assumed to have stoichiometric TiO_2 contents.

c) Model hydrous cumulate and residual liquid compositions produced at fL_2

For $fL_2 < 0.4$, the second hydrous cumulate assemblage is used to calculate the distribution coefficients.

The bulk Ds are calculated with hydrous assemblage #2 and the $D^{min/liq}$s reported in Table 2:

	Rb	Ba	Th	U	Nb	La	Ce	Pb	Pr	Sr	Nd	Sm	Zr[+]	Hf[+]	Eu	Ti*	Gd	Tb	Dy	Ho	Y	Er	Yb	Lu
Bulk D	0.23	0.42	0.32	0.19	0.77	0.63	0.75	0.11	0.95	0.48	1.21	1.58	0.33	0.55	1.52	1.60	1.71	1.63	1.57	1.50	1.43	1.28	1.07	1.00

* See previous comment on Ti calculation. [+] For Zr and Hf, the bulk Ds correspond to the assemblage without zircon; stoichiometric Zr and estimated Hf contents of zircon (see discussion in the main article) are used to estimate the effects of zircon fraction on the Zr and Hf concentrations in the liquid.

Composition of residual liquid at fL_2 (liquid L_2 in Fig. 5b):

	Rb	Ba	Th	U	Nb	La	Ce	Pb	Pr	Sr	Nd	Sm	Zr^{++}	Hf^{++}	Eu	Ti**	Gd	Tb	Dy	Ho	Y	Er	Yb	Lu
Residual Liquid at f L2 (ppm)	31.7	301	4.21	1.79	30.6	27.0	48.7	6.94	6.18	678	15.0	2.55	197	3.58	0.61	2606	1.78	0.31	2.00	0.38	9.2	0.79	0.69	0.08

** See previous comment on Ti liquid calculation. $^{++}$ Zr and Hf contents of the residual liquid are calculated in two steps. First, the Zr and Hf contents in zircon (Table 2) are subtracted from the initial liquid composition; second, the fractional crystallisation equation is used to calculate the effect of fractionally removing the remaining solid phases.

Model Hydrous cumulate composition produced between f = 0.4 and fL_2 (in this example fL_2 = 0.266):

	Rb	Ba	Th	U	Nb	La	Ce	Pb	Pr	Sr	Nd	Sm	Zr	Hf	Eu	Ti	Gd	Tb	Dy	Ho	Y	Er	Yb	Lu
Model hydrous cumulate (ppm)	6.2	112	1.18	0.29	22.3	15.7	34.7	0.61	5.8	288	19.0	4.59	86	2.34	1.03	9442	3.56	0.57	3.57	0.64	14.4	1.07	0.75	0.08

Amphibole composition in the model hydrous cumulate assemblage produced between f = 0.4 and fL_2:

	Rb	Ba	Th	U	Nb	La	Ce	Pb	Pr	Sr	Nd	Sm	Zr	Hf	Eu	Ti	Gd	Tb	Dy	Ho	Y	Er	Yb	Lu
Amphibole (ppm)	2.63	95	0.07	0.02	15.4	5.74	18.5	0.55	3.85	417	14.3	4.04	73.1	2.35	0.98	7405	3.44	0.57	3.60	0.66	14.9	1.12	0.77	0.09

d) Mean model hydrous metasomatic vein composition produced between fmax and fL_2

The composition of the hydrous cumulate assemblage produced between $f$max and fL_2 is calculated using the weighted averages of the cumulate phases produce from $f$max to f = 0.4 and from f = 0.4 to fL_2.

Model hydrous cumulate composition produced between $f$max and fL_2 (corresponding to hydrous cumulate assemblage plotted in Fig. 5c):

	Rb	Ba	Th	U	Nb	La	Ce	Pb	Pr	Sr	Nd	Sm	Zr	Hf	Eu	Ti	Gd	Tb	Dy	Ho	Y	Er	Yb	Lu
Model hydrous cumulate (ppm)	6.2	112	1.19	0.28	22.7	16.0	35.3	0.60	5.9	288	19.3	4.66	85	2.32	1.05	9837	3.61	0.58	3.61	0.65	14.6	1.09	0.76	0.08

The composition of amphibole in the hydrous cumulates produced between $f$max and fL_2 is calculated using the amphibole produced from $f$max to f = 0.4 and the amphibole produced from f = 0.4 to fL_2 weighted as function of their proportions in each interval.

Amphibole composition in the model hydrous cumulates produced between $f$max and fL_2 (corresponding to the amphibole composition plotted in Fig. 6):

	Rb	Ba	Th	U	Nb	La	Ce	Pb	Pr	Sr	Nd	Sm	Zr	Hf	Eu	Ti	Gd	Tb	Dy	Ho	Y	Er	Yb	Lu
Amphibole (ppm)	2.60	94	0.07	0.02	15.3	5.70	18.4	0.54	3.85	413	14.4	4.08	72.8	2.34	0.99	7804	3.49	0.58	3.63	0.67	15.0	1.13	0.77	0.09

Part 3 - Composition of model *metasomatized hydrous lithosphere* (hydrous metasomatic veins + cryptic enrichment in the peridotite surrounding the veins)

As indicated in the main text, we model the potential cryptic and modal metasomatic enrichment in peridotite associated with metasomatic veins by including phlogopite with the hydrous cumulates and trapping residual liquid in the peridotite surrounding the veins (see Fig. 4). The addition of trapped liquid to the hydrous cumulates yields the composition of *metasomatized hydrous lithosphere* plotted in Fig. 5d.

Fraction of residual liquid trapped in surrounding peridotite: 0.180 This parameter is free to vary from 0 to 0.3 but is linked to the proportion of phlogopite (in this example: 0.06) in the hydrous vein by a ratio of phlogopite to trapped residual liquid of 1 to 3.

The composition of *metasomatized hydrous lithosphere* is calculated using the proportions of trapped liquid (L_2) and hydrous cumulates (those produced between $f$max and fL_2).

Composition of the model *metasomatized hydrous lithosphere* (plotted in Fig. 5d):

	Rb	Ba	Th	U	Nb	La	Ce	Pb	Pr	Sr	Nd	Sm	Zr	Hf	Eu	Ti	Gd	Tb	Dy	Ho	Y	Er	Yb	Lu
Metasomatized hydrous lithosphere (ppm)	17.9	22	21.84	27.09	36.7	27.8	22.5	11.60	23.4	18	14.8	10.53	10	9.01	6.28	7	6.02	5.37	4.92	4.04	3.2	2.35	1.69	1.18